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RESONANCE STUDIES OF HYDROGEN ATOMS INTERACTING WITH VERY COLD SURFACES

March 1, 1981 - February 28, 1982

The President and Trustees
of Williams College
Williamstown, Massachusetts
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Experimental techniques and theoretical methods of analysis for the study of hydrogen atoms stored in containers having very cold surfaces are developed. The temperature dependence of the probability of adsorption and average time of adsorption of hydrogen atoms on frozen molecular hydrogen surfaces is measured. Recombination and other relaxations while adsorbed are studied. A high intensity state-selected hydrogen atom beam with thermal speeds at 6 K is developed and applied to atomic hydrogen maser development.

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ANNUAL SUMMARY REPORT

ONR Contract # N00014-80-C-0240

RESONANCE STUDIES OF HYDROGEN ATOMS INTERACTING WITH VERY COLD SURFACES

1. Principal Investigator: Dr. Stuart B. Crampton
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2. Contract Desription:

Techniques are developed for studying the probability that a hydrogen atom is adsorbed and the average time adsorbed when it collides with a liquid helium temperature molecular hydrogen surface. A theory is developed that relates the frequency and radiative life-time of a pulsed hyperfine resonance signal to the probability of adsorption and mean adsorption time per collision with the surface. Recombination of atoms while adsorbed and electron spin exchange collisions between atoms while adsorbed are studied. A liquid helium temperature state-selected hydrogen atom beam is developed and used to study the feasibiltiy of atomic hydrogen maser oscillation at liquid helium temperatures.

3. Scientific Problem

The energies of adsorption of hydrogen atoms by very cold surfaces, the durations of surface adsorptions, and the recombination and spin relaxation interactions between hydrogen atoms while adsorbed are important to current attempts to cool sufficient quantities of spin polarized hydrogen atoms in high magnetic fields to low enough temperatures to produce Bose-Einstein condensation in a gas, but these parameters are more precisely observable in low magnetic fields, as in these studies. The density of hydrogen atoms that can be stored in low magnetic fields in containers coated with inert surfaces is high enough and the relax-ation times while stored are long enough to offer attractive opportunities for precision measurements and for improved frequency metrology. The electron exchange collision relaxations and frequency shifts at very low temperatures offer sensitive tests of the fundamental interactions between colliding hydrogen atoms. Operation at very low temperatures offers potentially significant improvements of both short term and long term atomic hydrogen maser frequency stability.

4 Scientific and Technical Approach

These techniques are extensions to somewhat higher and lower temperatures of the techniques developed by this investigator in collaboration with T. J. Greytak, D. Kleppner, W. D. Phillips, A. Weinrib and D. A. Smith at MIT for studying H atoms interacting with H_2 surfaces at 4.2 K. In those studies, which were continued at Williams College after December 1978, H atoms produced in a 180 Mhz rf discharge cooled to 77K pass through a 1 cm diameter and 20 cm long glass tube to a 100 cm^3 storage bottle centered in a microwave cavity tuned to the 1.42 GHz hydrogen atom ground state hyperfine transition frequency. Tube and storage bottle are immersed in a liquid helium bath whose temperature is controlled by controlling the density of helium vapor over the bath. Frozen H_2 surfaces are carefully prepared on the insides of the glass surfaces. A short pulse of rf power resonant at the hyperfine frequency perturbs the thermal H level populations and produces a coherent superposition of states. Subsequent radiation by the atoms is approximately an exponentially damped cosine, whose frequency and decay rate can be linked theoretically to the probability α of adsorption and the mean time $\langle t_a \rangle$ of adsorption per surface collision. The amplitude of the signal measures the density of stored atoms, and the recovery of signal amplitudes following pulses that disturb the level populations probes the level population recovery rates. These level population recovery rates provide information about the probability of recombination while adsorbed and other relaxation processes affecting the adsorbed atoms. At temperatures that are high enough relative to the adsorption energy these recovery rates are sensitive to the electron spin exchange collisons between H atoms in the gas phase.

The state-selected H atom beam is formed by passing H atoms dissociated in the liquid nitrogen cooled discharge through a small copper cylinder in which they are thermalized to about 6 K. After leaving the copper "accomodator," atoms in the (F=1,m=0) hyperfine state are preferentially focused by a large bore hexapole magnet. State-selected H atom beam intensities of order 20 times those possible with room temperature sources are anticipated.

In order that frequency perturbations and contributions to the resonance linewidths due to surface adsorptions be as small as possible, the surface should be at a temperature that is as high as possible compared to the adsorption energy but not so high that H atoms collide with the atoms or molecules of the saturated vapor over the surface. For frozen molecular hydrogen surfaces the optimum temperature is expected to be about 6 K, and for frozen neon surfaces the optimum temperature is expected to be about 10 K. These temperatures are just above those accessible by simply increasing the density of helium vapor over the liquid helium bath. For liquid helium surfaces the optimum temperature has been shown by Walter Hardy and his colleagues at the University of British Columbia to be about 0.5 K. That temperature is too low to be reached by simply pumping on the liquid helium bath, but it is accessible with a ^{3}He-^{4}He refrigerator.

5. Progress

The original plan for achieving temperatures low enough for liquid helium surfaces was to use the large ^{3}He-^{4}He dilution refrigerator belonging to the MIT spin polarized hydrogen group in alternation with their own investigations of spin polarized hydrogen in high magnetic fields. As the work of this group and that group progressed, it became clear that scheduling posed insuperable problems. Consequently, we have begun the construction here of a ^{3}He refrigerator here. Part of the insert was designed and built during this past year by Professor Jim Eisenstein and his undergraduate research student. (Professor Eisenstein received his PhD from Berkeley in 1980 on the basis of investigations of the properties of liquid ^{3}He in the millikelvin temperature regime. He has continued his low temperature research since coming to Williams as Assistant Professor, and he is a source of much useful advice about properties and techniques.) In the meantime our own results for H adsorbed on frozen H_2 surfaces have suggested that frozen neon at its optimum temperature around 10 K should be a better storage surface than liquid helium at its optimum temperature around 0.5 K. Further work on the ^3He refrigerator has been suspended until the properties of H adsorption on neon have been tested.

The state-selected H atom beam has been designed, and a preliminary version has been tested. On the basis of its performance we have designed and are currently assmebling an improved version. We anticipate that this version will produce state-selected beam intensities of 6 K H atoms up to $5 \times 10^{14}\ s^{-1}$. The beam will be useful for studying H adsorption on H_2 and neon in ways that are not possible without it. For example, we plan to probe the very low density level population recovery rates, in order to determine whether H atoms recombine or exchange electron spins with atoms strongly bound in surface imperfections. The intensity will be high enough to produce self-excited hydrogen maser oscillation with H_2 surfaces between 5 K and 6 K and with neon surfaces between 8 K and 12 K. The maser oscillation characteristics will be used to probe the electron spin exchange relaxations and frequency shifts, as they have been at higher temperatures by this investigator and others.

We have developed a detailed analytic theory of the resonance signal of atoms perturbed while adsorbed during collisions with the surface of a spherical storage container, and we have applied it to the analysis of the temperature dependence of the mean time of adsorption of H atoms on H_2 surfaces between 3.2 K and 4.6 K. The reproducibility and the temperature dependences of $\langle t_a \rangle$ and α have encouraged us to try neon surfaces at around 10 K. Solid neon is similar to solid molecular hydrogen but has much lower polarizability. Preliminary measurements of the characteristics of H storage using neon surfaces will determine whether neon surafces at 10 K or liquid helium surfaces at 0.5 K offer the best prospects for hydrogen maser development.

6. Publications

We have submitted our results for H adsorbed on H_2 to Physical Review B as "Temperature Dependence of Hydrogen Atom Adsorption on Molecular Hydrogen Surfaces," by S. B. Crampton, J. J. Krupczak and S. P. Souza. It has been favorably reviewed but not yet published. This investigator gave an invited paper at the 3rd International

Symposium on Frequency Standards and Metrology held at Aussois, France, in October 1981. The paper was coauthored with J. J. Krupczak and S. P. Souza, and it will be published in a supplement to J. Physique (Paris) under the title "Progress of the State-Selected Beam Low Temperature Hydrogen Maser." These publications do cite ONR support.

7. Extenuating Circumstances

Originally, we had proposed that, after completing our studies of H adsorbed on frozen H_2 surfaces, we would perform similar studies of H adsorbed on liquid helium surfaces below 1 K. However, while our work with H_2 surfaces was still in progress, Walter Hardy and his colleagues at the University of British Columbia measured most of the parameters needed to evaluate the likely performance of an atomic hydrogen maser using liquid helium storage surfaces. In particular, they found that at the optimum temperature at which the hydrogen atom frequency is first order independent of temperature the mean free path of the hydrogen atoms in the saturated helium vapor is appreciably less than reasonable apparatus dimensions. Collisions with the vapor introduce many potential instabilities to hydrogen maser operation with liquid helium surfaces. Consequently, we have redirected our efforts to the study of neon storage surfaces. Neon has much lower polarizability per unit mass than helium, so that the vapor pressure should be still quite low at temperatures at which perturbations due to adsorption are also low. Of course, solid neon is not a self-healing superfluid, as liquid helium is at 0.5 K, and the reproducibility of the solid neon surfaces will be an important issue. Our results for H_2 surfaces are very encouraging on this point.

Other than this redirection to neon surfaces, the work has been progressing on a reasonable schedule. The results to date are highly relevant to work elsewhere on spin polarized hydrogen, to any kind of resonance studies of hydrogen atoms interacting with cold surfaces, and to applications of the state-selected beam to other problems in physics.

8. Unspent Funds

We have already spent or have committed to be spent by February 28, 1982, about 98% of the funds available until then, and we do not anticipate any surplus.

9. Graduate Student Degrees

There is no graduate program in physics at Williams College. One undergraduate student who has participated in this work for two years and who plans to write an honors thesis based on his work does plan to go on to gradaute school next year. Another former Williams student, who has been working full time on this work since graduating in June 1980 and whose name appears on our papers, also plans to go on to graduate school in physics next year.

10. Other Federal Grant and Contract Support

Contract # 955441 from the Jet Propulaion Laboratory provided about $22000 per year of parallel support for this work for two years through October 1982. It has not and will not be renewed. Grant # PHY79-10967 A01 from the NSF has provided parallel support for this work in the amount of $54310 since November 1979. A renewal proposal has been submitted but has not been acted upon yet.

Stuart B. Crampton
Principal Investogator